LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

UCRL-PROC-210585

Density and Tensile Properties Changed by Aging Plutonium

B. W. Chung, B. W. Choi, S. R. Thompson, C. H. Woods, D. J. Hopkins, B. B. Ebbinghaus

March 16, 2005

26th Compatibility, Aging, and Stockpile Stewardship Conference
Aiken, SC, United States
April 26, 2005 through April 28, 2005

Density and Tensile Properties Changed by Aging Plutonium

B.W. Chung, B.W. Choi, S.R. Thompson, C.H. Woods, D.J. Hopkins, and B.B. Ebbinghaus

University of California, Lawrence Livermore National Laboratory, P.O. Box 808, Livermore, CA 94551

Abstract

We present volume, density, and tensile property change observed from both naturally and accelerated aged plutonium alloys. Accelerated alloys are plutonium alloys with a fraction of Pu-238 to accelerate the aging process by approximately 18 times the rate of unaged weapons-grade plutonium. After thirty-five equivalent years of aging on accelerated alloys, the dilatometry shows the samples at 35°C have swelled in volume by 0.12 to 0.14 % and now exhibit a near linear volume increase due to helium in-growth while showing possible surface effects on samples at 50°C and 65°C. The engineering stress of the accelerated alloy at 18 equivalent years increased significantly compared to at 4.5 equivalent years.

Introduction

Plutonium, because of its radioactive nature, ages from the "inside out" by means of self-irradiation damage and thus produces Frankel-type defects (vacancies and self-interstitial atoms) and defect clusters. The self-radiation damage in Plutonium-239 occurs mainly by alpha-particle decay, where most of the damage comes from the U-235 recoil nucleus. The defects resulting from the residual lattice damage and helium in-growth could result in microstructural and physical property changes. Because these self-irradiation effects would normally require decades to measure, a fraction (7.5 wt%) of Pu-238 is added to the reference plutonium alloy thus accelerating the aging process by approximately 18 times. By monitoring the properties of the Pu-238 spiked alloy over a period of about 3.5 years, the properties of plutonium can be projected for periods up to about 60 years. This paper presents density, volume, and tensile changes observed from both naturally and accelerated aged plutonium alloys.

Experimental

Each dilatometer unit consists of a small vacuum controlled-atmosphere sample chamber fitted with three linear variable differential transducers (LVDTs). An LVDT measures minute changes, 0.1 micron or less, in the position of a push-rod by monitoring changes in the inductance of a detector coil. In the current design, the detector coil is placed outside of the sample chamber. Two different lengths (2 and 3 cm) of alloy specimens are used to differentiate between surface oxidation and volumetric swelling in the materials. These alloys have nominal gallium concentration of 0.5 weight percentage. These specimens are placed in the copper well located inside the dilatometer system. A reference low thermal expansion glass (Zerodur) is also placed in the copper well to monitor the stability of the dilatometry system. Each dilatometer system was then maintained at a separate temperature, i.e., at 35°C, 50°C, or 65°C.

The immersion density equipment closely matches a design used by Bowman et al.[1] and uses about 200 ml of Fluorinert Electronic Liquid FC-43 as the immersion fluid. Prior to use, the system is calibrated using NIST glass (SRM 1827A). Because the ^{238}Pu-spiked alloys generate heat, a test sample is left overnight in the immersion bath to allow the temperature of the bath to stabilize and the measurements to be reproducible. A correction needs to be applied to the measured density to compensate for the heat generated by the ^{238}Pu-spiked sample. A 5 g sample of 7.5 wt% ^{238}Pu- spiked alloy produces 225 mW of power. A series of power generating samples were made of tantalum heated with tungsten heating wire to simulate self heating of plutonium, and powers ranging from 0 to 1030 mW were run in the immersion density unit. These tests showed that for 225 mW of power, a correction of + 0.06 g/cm^3 needs to be applied to the measured density. Specimens for the immersion density are identical in composition to the dilatometry specimen.

Each tensile test specimen was dimensionally inspected for the gage diameter and its length (GL), and loaded into a specially designed fixture for the tensile test. With a 0.24 inch GL extensiometer for the strain measurement, all testing was performed at crosshead speed of 0.05 inch/min, so that the ultimate strain rate was about 3.5 x 10^{-3}/sec. Aluminum specimens of known tensile strength were tested to verify the equipment integrity before and after each test specimen. The load and displacement data was recorded on a computer using the Instron Series IX software package in conjunction with an Instron Model 4444 test machine. Data was recorded from preloading until failure. Plutonium specimens for the tensile test have nominal gallium concentration of 1 weight percentage.

Results and Discussion

The volume change (ΔV) normalized with the initial volume (V) of each spiked alloys at 35, 50, and 65°C is shown in Figure 1. The time is represented as an equivalent time (in year) obtained by multiplying the measurement time by the accelerating factor of 18.59. This accelerated factor is obtained by the decay rate of spiked alloy normalized by reference alloy. This factor will decrease as the material ages due primarily to in-growth of ^{241}Am in the reference alloy. Each dilatometer contains a pair of long (3cm) and short (2cm) length specimens. The $\Delta V/V$ of the sample is obtained with the relation $\Delta V/V = 3\ \Delta L/L$ where $\Delta L/L$ represents the measured sample length change (ΔL) normalized with the initial length (L). As plotted in Figure 1, all the spiked alloys have increased in volume significantly. During the early stage of measurement, samples at 35, 50, and 65°C storage temperatures increased in volume as a result of self-irradiation and follows the inverse exponential-type of expansion on dose or time during initial stage of aging. This behavior has been attributed to the lattice damaged caused by the radiation damage. After the initial expansion, the volume change exhibits significantly lower rate of increase and at a near linear expansion behavior attributed to the helium in-growth mechanism. The difference in $\Delta V/V$ values for short (s) and long (l) specimens is believed to result mainly from surface effects (i.e. oxidation and/or interfacial reaction) in the specimens. There is a trend in $\Delta V/V$ curves with temperature since samples expanded more with an increase in temperature. The short specimens at 65°C and 50°C give higher volume change values compared to the long specimens. At 35°C, both 2 and

3 cm length samples (open and closed squares) show the same ΔV/V indicating minimal surface contribution, and the measured volume (length) change is by volumetric swelling.

The curves for 35°C are quite accurately represented by the combination of exponential and linear growth equations of the form[2]

$$\Delta V/V = A\,[1-\exp(-Bt)] + Ct \qquad (1)$$

where A, B, and C are constants and t is the time in years. For 50°C and 65°C, a fit based on a linear relation was performed at the saturation (or linear) region to obtain the slope C. The He/vacancy association ratios are calculated using the slope (C). This ratio describes the volume expansion induced by the formation of the helium bubbles in Pu metal. The average ratio extracted from the curve fit is approximately 2.5.

Comparison of density change observed in aged reference alloys to the 35°C dilatometry data is shown in Figure 2. The immersion density measurement on the reference (weapons-grade Pu) and ^{238}Pu-spiked alloys showed initial densities of 15.795 and 15.78 g/cc, respectively. The ages of reference samples range from 0.2 to 21 years. The immersion density measurement shows large variation in density values. One reason might be differences in processing of these materials. The initial density value for the dilatometry data is set to 15.795 g/cc to compare to the reference alloys. The density value from the dilatometry corresponds well to the immersion density.

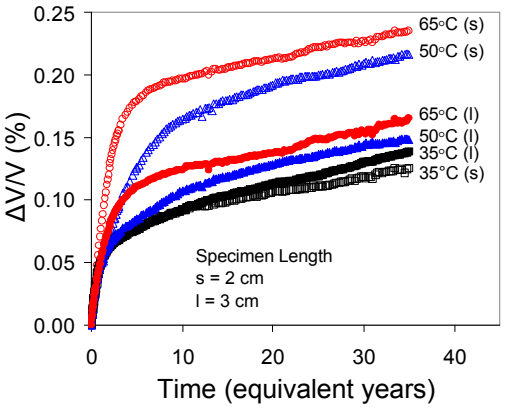

Figure 1. The normalized volume changes for spiked alloys tested under different temperatures. 2 and 3 cm length samples are described as "s" and "l".

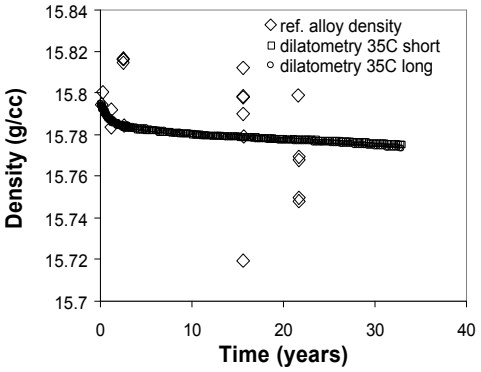

Figure 2. Comparison of density change calculated from dilatometry (35°C) to immersion densities of reference alloys.

Figure 3 shows the stress-strain curves of two 27 years old reference alloy samples and two spiked alloy samples at 4.5 and 18 equivalent years, respectively. As can be seen from the data, the engineering stress of the spiked alloy at 18 equivalent years increased significantly compared to at 4.5 equivalent years. The aged (18 equiv. years) spiked alloy has increased in the stress to about the same as the 27 years old reference alloy but shows a difference in the total elongation between the two alloys.

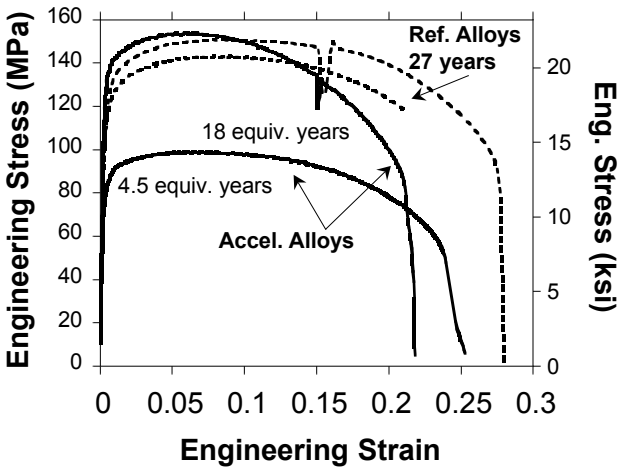

Figure 3. Engineering stress versus Engineering strain for two reference alloy samples and one spiked alloy sample at 4.5 and 18 equivalent years.

Summary and Conclusion

Dilatometry, immersion density, and tensile measurements are underway on ^{238}Pu-spiked alloys and reference alloys. Current measurements show the volume (length) expansion from the accumulation of residual lattice damage and helium in-growth. Dilatometry measurements show volumetric changes (expansion) caused by self-radiation damage. The average He/vacancy ratio was extracted to be about 2.5. Further experiments are needed to determine how oxidation or interfacial reaction affects the measurement for longer term expansion rates. Density changes calculated from the dilatometry corresponds well to the immersion density values during current time period. Tensile measurements Pu specimens show increased engineering stress with aging.

Acknowledgment

This work was performed under the auspices of the U.S. Department of Energy by the University of California, Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

References

1. H. A. Bowman, et al., *J. of Res. Nat. Bur. Stand*., 71C (3), 179 (1967).
2. B.W. Chung, S.R. Thompson, C.H. Woods, D.J. Hopkins, W.H. Gourdin, and B.B. Ebbinghaus, submitted to J. Nucl. Mater.